ARKIV FÖR FYSIK Band 12 nr 20

Read 12 September 1956

Gamma radiation from some Swedish foodstuffs

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With 5 figures in the text

Summary

The γ radiation from powdered milk, beef, bone from cattle, and vegetables, collected and preserved in the years 1952–56, has been investigated in May–September 1956 by means of two high-pressure ion chamber apparatuses placed in a low-background laboratory. In every case a significant increase has been observed which seems to originate from fallout from atomic bomb tests. Investigations of the γ radiation from children have been repeated, but no increase could be observed in comparison with earlier tested subjects in approximately the same age and weight classes (101 subjects, 1954; 79 subjects, 1956).

(Some further measurements of γ radiation from powdered milk, beef, bone, and thyroid from grazing cattle have been added in the proof.)

Two apparatuses consisting of cylindrically arranged high-pressure ion chambers (diameter of sample container approx. 1 m, total chamber volume approx. 3 m^3, 20 kg N_2 and CO_2) placed in a low-background laboratory in a rock near the city of Stockholm, and protected in all directions by 1 m of water and more than 50 m of granite, have been described in earlier communications [1, 2, 3]. In these, the results of measurements on 306 persons who have had no known contact with other than natural radioactive material of common concentration were also reported. The γ radiation of human subjects is caused mainly by the K^{40} content of the body, as has been shown by Burch and Spiers [4].

The same instruments have, in May–July 1956, been used for determination of the possible amount of γ-radiating fission products in powdered milk and beef preserved in the years 1953 to 1956. The potassium content of the powdered milk was determined by the Institute of Radiophysics and the Department of Clinical Biochemistry at Karolinska Sjukhuset, entirely independently. The absorption of the radiation from K^{40} and Cs^{137} was measured by mixing 500 g of potassium chloride and 0.1 μC of Cs^{137}, respectively, in the milk samples, each of which weighed 10 kg.

The results of these measurements will be seen from the graph in Fig. 1. An increased γ radiation is obvious, which cannot have been caused by differences in the potassium content. If this increase is due entirely to Cs^{137}, the concentration in the natural milk for the samples from 1956 can be calculated at about 1×10^{-4} μC per liter of milk, that is, 5×10^{-5} of the maximum permissible concentration. Because of the comparatively low age of the last samples it seems, however, likely that other fission products may have played the predominant role.

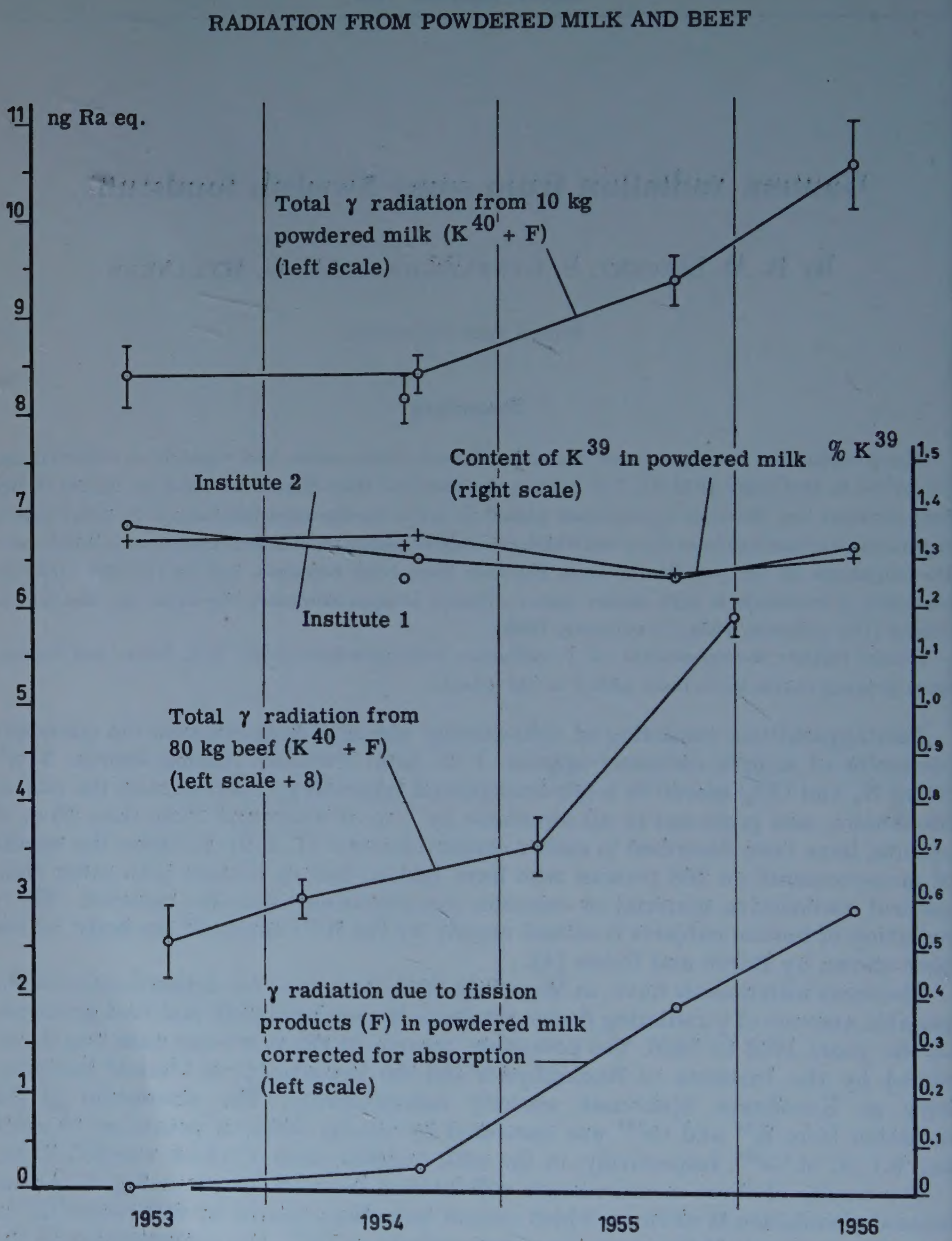


Fig. 1. All the measurements were carried out between May 18–July 9, 1956. The time is plotted on the graph corresponding to the preservation date of the samples. The standard errors of the observed radiation values are given.

Beef from the years 1953–55 was examined too, each sample consisting of 80 kg of meat. The results are also shown in Fig. 1. Since the potassium content is probably the same in all these samples, the higher γ radiation found in the last year might be attributable, here too, to an increase in fission products.

In August–September 1956 further investigations were carried out, the following results being obtained:

Table 1.

Sample	Date of sample	Average age of sample, months	Radiation* ng Ra eq.	Average age of sample, months	Radiation* ng Ra eq.	Potassium content %
Powdered milk 10 kg (natural milk 87 kg)	1953 June	36	8.1	38	8.5	1.36
	1956 June	< 1	10.5	2–3	10.1	1.31
	Difference		2.4		1.6	
20 kg	1953 June	38	16.6	38	16.6	1.35
	1956 Sept.	< ¼	35.6	¼–½	30.6	1.27
	Difference		19.0		14.0	
Beef 80 kg	1953	36**	10.6	37½**	10.9	—
	1955 Dec.	7**	13.9	8½**	14.4	—
	Difference		3.3		3.5	
Bone from cattle 40 kg (boiled)	1953	36	9.4			0.22
	1956 June	3	14.4			0.20
	Difference		5.0			
Oats 100 kg (50 kg)	1952	48	27.0	48	(13.8)	0.39
	1954	24	29.2	24	(15.7)	0.43
	Difference		2.2		(1.9)	
Apples 45%, Hips 55% (dried) 80 kg (40 kg)	1953	36	10.3	36	(5.5)	0.27
	1955	12	12.1	12	(6.7)	0.29
	Difference		1.8		(1.2)	

* Corrected for differences in the potassium content. Figures in parentheses refer to samples of half the weight, 50 kg and 40 kg respectively.

** These figures are uncertain, part of the preserved meat possibly being several months older.

From Table 1 it is obvious that all the substances investigated show an increased γ radiation which cannot be attributed to any natural radioactive elements all the radiation values being determined from 8 observations with a standard error of less than 4.5 %.

The γ radiation of the samples taken in 1952 and 1953 can probably be regarded as entirely due to K^{40}, because of the comparatively small amounts of fallout in Sweden before the end of 1953. A calculation of the γ radiation from the potassium content in these samples gives values which seem to be in good agreement with the radiation observed.

Table 2.

(Ru 106 produced from Pu; all other elements from U and Pu.)

Estimated duration of appreciable contribution to observed γ radiation, counted from explosion date [5]	Element	$T_{\frac{1}{2}}$	Energy of γ-rays MeV	Critical organ [6]	Effective half-life [6]	M.P.C.* in water μC/cm^3 [6]	M.P. total body burden μC [6]
	I 131	8 d.	0.28–0.64	Thyroid	7.5 d.	6×10^{-5}	0.6
10–60 d.	Ba 140	12.8 d.	0.16–0.54	Bone	12 d.	0.3×10^{-3}	1
	La 140	40 h.	0.33–2.5	Bone	1.6 d.		
20–90 d.	Ru 103	42 d.	0.50	Kidney			
30–120 d.	Ce 141	30 d.	0.15	Bone			
20 d.–1.5 y.	Zr 95	65 d.	0.72–0.75	Bone	48 d.	0.6×10^{-3}	10
	Nb 95	35 d.	0.75	Bone	21 d.		
1–5 y.	Ce 144	275 d.	0.13	Bone	180 d.	0.1×10^{-3}	1
	Pr 144	17.5 m.	1.48–2.18	Bone			
1–6 y.	Ru 106	1 y.	—	Kidney	19 d.	0.1×10^{-3}	4
	Rh 106	30 s.	0.52–1.54				
> 2 y.	Cs 137	33 y.	—	Muscle	17 d.	2×10^{-3}	100
	Ba 137	2.6 m .	0.66	(Bone)			

* Based on a maximum permissible dose rate to a portion of the gastrointestinal tract.

The difference in the portion of γ radiation from fission products when measuring samples of vegetables of different weight is caused by the average energy being lower for the fission products than for K^{40} (1.54 MeV).

The fission products which may be of particular interest for the explanation of the γ radiation observed are collected in Table 2.

It may be noted that Miller and Marinelli in a recent publication [7] reported that some but not all meats and milk powders were found to give positive results, whereas drinking water, vegetables and scallops proved to be negative as to the photopeak of cesium observed by means of a scintillation counter.

The tests described here indicate that vegetables as well are contaminated. This conclusion seems to be justified, since the determinations of the potassium content of the substances investigated in no case showed any differences between old and new samples of such a magnitude that the increased γ radiation could be explained by γ radiation from K^{40}.

Although the quantities observed would probably be too small to cause a detectable increase in the γ radiation from human subjects, series of children were investigated once more, and the results compared with the earlier measurements referred to in the beginning of this communication. As seen from Table 3 the two sets of observations (1954 and 1956) are well suited for such a comparison. No significant change could be detected, however, in the average γ radiation.

Further measurements in September 1956 to January 1957 have given the following results, which may be added in the proof.

From the middle of September to the last part of November samples of powdered milk, beef, bone and thyroids from grazing cattle were periodicallly

Table 3.

Body weight kg . . .	< 25	25.1–35.0		35.1–45.0		45.1–55.0	
	F	M	F	M	F	M	F
1954							
Number of subjects .	6	18	18	20	17	10	12
Average weight (a) .	22.2	30.0	30.4	40.4	40.6	49.5	49.6
Average age	8.1	9.4	9.1	12.2	11.9	13.3	13.7
γ rad. in ng Ra eq.							
min.	2.23	2.80	2.05	2.94	1.99	4.41	3.39
max.	4.12	4.95	4.45	6.62	6.45	7.32	7.18
average (b)	3.12	3.74	3.35	4.87	4.22	5.81	5.22
10 b/a	1.41	1.25	1.10	1.21	1.04	1.17	1.05
Standard error . . .	0.077	0.057	0.050	0.052	0.072	0.050	0.065
1956							
Number of subjects .	2	13	16	27	10	5	6
Average weight (a) .	21.5	31.2	31.6	40.3	39.7	50.8	47.5
Average age	10.0	10.5	10.4	12.3	12.6	13.2	13.3
γ rad. in ng Ra eq.							
min.	2.57	2.70	2.20	2.69	3.03	5.68	3.66
max.	2.63	5.35	5.17	6.39	7.01	6.25	6.45
average (b)	2.60	4.10	3.67	4.55	4.47	6.09	4.50
10 b/a	1.21	1.31	1.16	1.13	1.13	1.20	0.95
Standard error . . .	0.014	0.083	0.067	0.042	0.091	0.024	0.053

investigated. The samples were standardized to 20 kg of powdered milk, 45 kg of beef, 20 kg of bone and 1 kg of thyroid, the three last-named samples being preserved in alcohol in polyethene bottles of 2×25, 30, and 2 ½ liters respectively. The thyroids were measured in the centre of a spherical high pressure ion chamber; the other samples in the apparatus already mentioned, the response of which had been checked for some of the isotopes in Table 2. The values in 10^{-9} g Ra eq. corresponding to 1 μC under the conditions used will be seen from Table 4.

The γ radiation from samples of various dates is shown in Figs. 2–5.

Table 4.

Isotope . . . . Quantity	Ru 106 1 μC	I 131 1 μC	Cs 137 1 μC	Sr 90 1μ C
Measured value in ng Ra eq.:				
No extra absorption	138 (100 %)	240 (100 %)	363 (100 %)	2.2 (100 %) in 50 g water
In centre of cylinder (diam. 35 cm, length 55 cm) containing 50 kg water	53 (38 %)	79 (33 %)	139 (39 %)	
Homogeneously distributed in cylinder, 50 kg water	66 (48 %)	116 (48 %)	193 (53 %)	1.4 (64 %) in 20 kg bone surrounded by 12 kg alc.

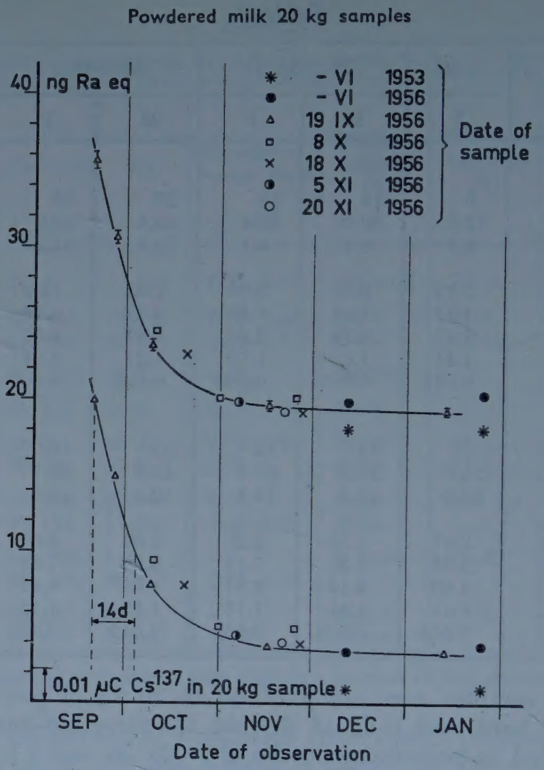


Fig. 2. The upper curve indicates the values observed and the lower curve the same values corrected for the K^{40} radiation based on determinations of the potassium content of the samples.

The decay curve for powdered milk in Fig. 2 indicates that a nuclear explosion has taken place in September. The content of potassium was determined for each sample, showing a general tendency to decrease from summer to fall.

The samples from June 1953 and 1956 were measured again in December 1956 January 1957. The difference between the last measurements of the 1953 and 1956 samples may possibly have been caused mainly by Cs^{137}. If so, the concentration in natural milk of Cs^{137} can be calculated at 0.06 $\mu\mu$C per ml of milk, corresponding to about 50 $\mu\mu$C Cs^{137} per g potassium. Some of the activity observed may, however, still have been due to fission products of shorter half-life (see Table 2). The calculated value consequently must be considered as a maximum estimation.

Most of the fallout of mixed fission products seems to have occurred in September or possibly also in early October, as the samples of later dates gave values close to or slightly above the decay curve from the sample of September 19.

The values in Fig. 3 which were obtained from beef do not permit any definite conclusions other than that the mixed fission products apparently are not taken up in any appreciable amount by muscle tissue. The results shown in Fig. 1 and Table 1 were not verifiable.

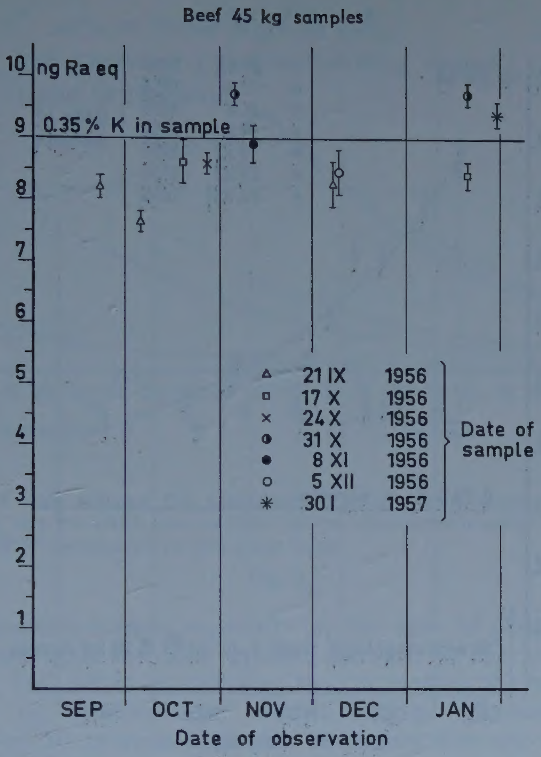


Fig. 3. The K^{40} radiation level indicated was determined by means of direct measurements of the potassium content of the samples and radiation measurements carried out by adding known amounts of potassium to one sample. The level is to be regarded as an approximate average.

The γ radiation from mixed fission products in bone, as will be seen from Fig. 4, shows a curve quite different from the curve for milk. The decay during the first month was probably caused chiefly by $Ba^{140}+La^{140}$ and $Zr^{95}+Nb^{95}$, and the values of the subsequent samples indicate that equilibrium is very soon reached between uptake and excretion. Dr. Bo Lindell at the Institute of Radiophysics has kindly made a determination of the amount of Sr^{90} in our last bone samples and has found about 1 $\mu\mu$C Sr^{90} pr g Ca that is 0.1 $\mu\mu$C Sr^{90} per g natural bone. Consequently the "Bremsstrahlung" from Sr^{90} in the bone samples gives a contribution which corresponds to less than 0.01 ng Ra eq. and therefore is much too small to be detected with the methods used in our determinations.

The determinations of I^{131} in the thyroids are collected in Fig. 5, indicating that the maximum of the uptake occurred in September and early October, which is in fairly good agreement with the results obtained from the milk and bone samples.

It may be noted here that the values in the curves of Figs. 2–5 have been subject to minor corrections because of some variations in the periodical calibration of the instruments and thus do not agree in detail with some earlier given data.

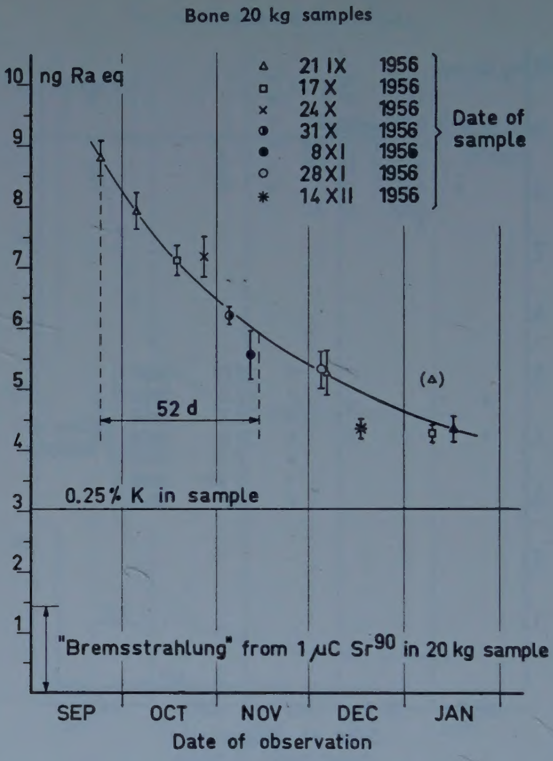
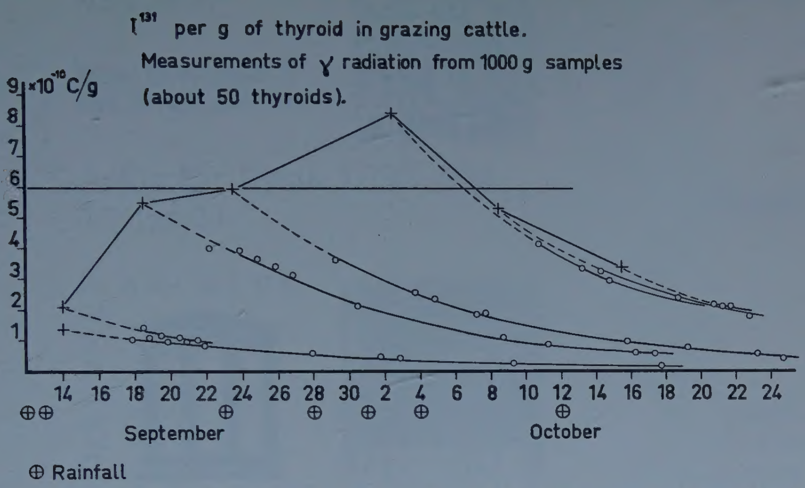


Fig. 4. The K^{40} radiation level indicated was determined by means of direct measurements of the potassium content of the samples and radiation measurements carried out by adding known amounts of potassium to one sample. The level is to be regarded as an approximate average. The value in parentheses was obtained from a sample in a container which, due to certain circumstances, was slightly contaminated on the outside. The lower value was obtained with a new container.

If the probable dose in the bone and thyroid is estimated on the basis of boneseakers (including Sr^{89}) and of I^{131} respectively, we arrive at average doses of the order of 1 mrad in bone and 200 mrad in thyroid over a period of one month. Since the variations of γ radiation from mixed fission products in the fallout have proved to be very large in Sweden [8], it seems likely that in some parts of the country doses of at least 10 times the calculated ones may have occurred. This would mean, for bone, doses of at least the same order of magnitude as, and for thyroid 200 times more than, the natural radiation during one month. If we assume the Sr^{90} concentration to be 5 $\mu\mu$C Sr^{90} per g Ca (5 times the observed value which is mainly due to earlier nuclear weapon tests), the average skeletal dose caused by Sr^{90} in one month would be less than one-tenth of the natural radiation, that is, 10 times lower than the maximum values that might have occurred in Sweden due to mixed fission products.

The maximum dose rate caused by a certain amount of radioactive material deposited in the skeleton may be dependent, furthermore, on the time during



The dotted lines indicate the extrapolation of the decay curves observed back to the last day on which the majority of the cattle were grazing. The straight line at $6 \cdot 10^{-10}$ corresponds to 0.05 r per week.

Fig. 5.

which the incorporation occurs, especially in the case of children. The hot spots in the bone change with the bone growth and are, in the long run, subject to a dilution effect. This effect contributes too, in making the maximum dose rate from Sr^{90} taken up by the bone over a long period considerably smaller—perhaps a factor of 10 or more—than the maximum dose rate from mixed fission products of short half-life taken up during a short period.

Accordingly, even at great distances from a nuclear weapon test area the mixed fission products of short and medium half-life including I^{131} may cause, sometimes for several months, a very much higher dose rate than the isotopes of long half-life. Since we do not know if there are thresholds in the biologic action of small doses of ionizing radiations, causing an increased effect of the higher dose rates in comparison with lower ones, it therefore seems advisable to study more carefully than hitherto the world distribution of mixed fission products during the first few months after nuclear weapon tests.

These investigations have been supported by the *Swedish Atomic Committee* and the *Knut and Alice Wallenberg Foundation.*

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Tryckt den 30 augusti 1957

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